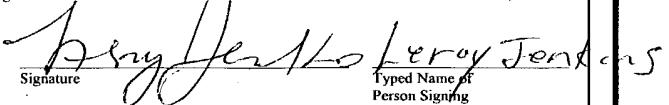
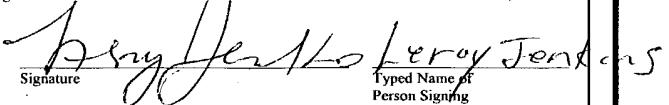
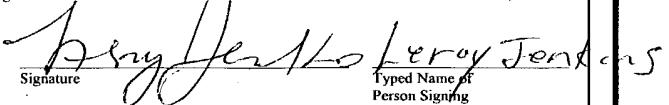
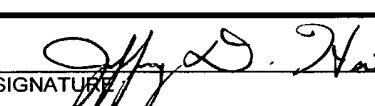


SUBSTITUTE FORM PTO-1390 U S DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE		ATTORNEY'S DOCKET NUMBER 13777-002001								
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371		U.S. APPLICATION NO. (If Known, see 37 CFR 15) 10/019471								
INTERNATIONAL APPLICATION NO. PCT/ZA00/00120	INTERNATIONAL FILING DATE 6 July 2000	PRIORITY DATE CLAIMED 6 July 1999								
TITLE OF INVENTION HIGH TEMPERATURE METATHESIS PROCESS										
APPLICANT(S) FOR DO/EO/US Jan Mattheus Botha, Alta Spamer, Muzikayise Mthokozi Justice Mbatha, Bongani Simon Nkosi, Jan Petrus Karel Reynhardt, Kelvin Stephen Jacobus and Gavin Wyatt Schwikkard										
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:										
<ol style="list-style-type: none"> <input checked="" type="checkbox"/> This is a FIRST submission of items concerning a filing under 35 U.S.C. 371. <input type="checkbox"/> This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371. <input type="checkbox"/> This is an express request to promptly begin national examination procedures (35 U.S.C. 371(f)). <input type="checkbox"/> The US has been elected by the expiration of 19 months from the priority date (PCT Article 31). <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2)) <ol style="list-style-type: none"> a. <input checked="" type="checkbox"/> is attached hereto (required only if not communicated by the International Bureau). b. <input type="checkbox"/> has been communicated by the International Bureau. c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US). <input type="checkbox"/> An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)). <input checked="" type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)) <ol style="list-style-type: none"> a. <input type="checkbox"/> are attached hereto (required only if not communicated by the International Bureau). b. <input type="checkbox"/> have been communicated by the International Bureau. c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired. d. <input checked="" type="checkbox"/> have not been made and will not be made. <input type="checkbox"/> An English language translation of amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)). <input type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)). <input type="checkbox"/> An English language translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)). 										
Items 11 to 16 below concern other documents or information included:										
<ol style="list-style-type: none"> <input checked="" type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included. <input checked="" type="checkbox"/> A FIRST preliminary amendment. <input type="checkbox"/> A SECOND or SUBSEQUENT preliminary amendment. <input type="checkbox"/> A substitute specification. <input type="checkbox"/> A change of power of attorney and/or address letter. <input type="checkbox"/> Other items or information: 										
<input type="checkbox"/> <input type="checkbox"/> <input type="checkbox"/> <input type="checkbox"/> <input type="checkbox"/>										
<table border="1" style="width: 100%; border-collapse: collapse;"> <tr> <td style="width: 30%; text-align: center; padding: 2px;">CERTIFICATE OF MAILING BY EXPRESS MAIL</td> <td style="width: 40%; text-align: center; padding: 2px;">Express Mail Label No <u>EL485781779US</u></td> </tr> <tr> <td colspan="2" style="text-align: center; padding: 2px;">I hereby certify under 37 CFR §1.10 that this correspondence is being deposited with the United States Postal Service as Express Mail Post Office to Addressee with sufficient postage on the date indicated below and is addressed to the Commissioner for Patents, Washington, DC 20231</td> </tr> <tr> <td style="text-align: center; padding: 2px;">12-28-01</td> <td style="text-align: center; padding: 2px;">Signature </td> </tr> <tr> <td style="text-align: center; padding: 2px;">Date of Deposit</td> <td style="text-align: center; padding: 2px;">Typed Name of Person Signing</td> </tr> </table>			CERTIFICATE OF MAILING BY EXPRESS MAIL	Express Mail Label No <u>EL485781779US</u>	I hereby certify under 37 CFR §1.10 that this correspondence is being deposited with the United States Postal Service as Express Mail Post Office to Addressee with sufficient postage on the date indicated below and is addressed to the Commissioner for Patents, Washington, DC 20231		12-28-01	Signature 	Date of Deposit	Typed Name of Person Signing
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12-28-01	Signature 									
Date of Deposit	Typed Name of Person Signing									

U.S. APPLICATION NO. (IF KNOWN) <u>U 194</u>		INTERNATIONAL APPLICATION NO. PCT/ZA00/00120	ATTORNEY'S DOCKET NUMBER 13777-002001
17. <input checked="" type="checkbox"/> The following fees are submitted:		CALCULATIONS PTO USE ONLY	
Basic National Fee (37 CFR 1.492(a)(1)- (5)):			
Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO \$1040			
International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO \$890			
International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO \$740			
International preliminary examination fee paid to USPTO (37 CFR 1.482) but all claims did not satisfy provisions of PCT Article 33(1)-(4)..... \$710			
International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(1)-(4)..... \$100			
ENTER APPROPRIATE BASIC FEE AMOUNT =			
\$890.00			
Surcharge of \$130 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)).		\$0.00	
Claims	Number Filed	Number Extra	Rate
Total Claims	21 - 20 =	1	x \$18
Independent Claims	2 - 3 =	0	x \$84
MULTIPLE DEPENDENT CLAIMS(S) (if applicable)		+ \$280	
TOTAL OF ABOVE CALCULATIONS =		\$1,008.00	
<input type="checkbox"/> Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above are reduced by 1/2.		\$0.00	
SUBTOTAL =		\$1,008.00	
Processing fee of \$130 for furnishing the English Translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f))		\$0.00	
TOTAL NATIONAL FEE =		\$1,008.00	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property +		\$0.00	
TOTAL FEES ENCLOSED =		\$1,008.00	
		Amount to be refunded:	\$
		Charged:	\$
a. <input checked="" type="checkbox"/> A check in the amount of \$1,008.00 to cover the above fees is enclosed.			
b. <input type="checkbox"/> Please charge my Deposit Account No. 06-1050 in the amount of \$0.00 to cover the above fees. A duplicate copy of this sheet is enclosed.			
c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 06-1050. A duplicate copy of this sheet is enclosed.			
NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b) must be filed and granted to restore the application to pending status.			
SEND ALL CORRESPONDENCE TO:			
Y. Rocky Tsao FISH & RICHARDSON P.C. 225 Franklin Street Boston, Massachusetts 02110-2804 (617) 542-5070 phone (617) 542-8906 facsimile		 SIGNATURE Jeffrey D. Hsi for Y. Rocky Tsao NAME 40,024 34,053 REGISTRATION NUMBER	

10/019471

Attorney's Docket No.: 13777-002001 / USP/F221/JL/sg

531 Rec'd PCT/TT 28 DEC 2001

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant : Jan Mattheus Botha, et al.
Serial No. : Unassigned
Filed : Herewith
Title : HIGH TEMPERATURE METATHESIS PROCESS

BOX PCT

Commissioner for Patents
Washington, D.C. 20231

PRELIMINARY AMENDMENT

Prior to examination, please amend the application as follows:

In the claims:

Cancel claims 1-34.

Add claims 35-55.

35. (New) A metathesis process for the metathesis of Fischer-Tropsch olefins in the C₅ to C₁₅ range, said metathesis process including the step of subjecting a Fischer-Tropsch olefin feedstock in the C₅ to C₁₅ range to a metathesis catalyst at a temperature of from 300°C to 600°C and a pressure of from 1 to 30 Bar, said olefin feedstock including mono-methyl branched olefins.

36. The metathesis process as claimed in claim 35, wherein said process is carried out at a temperature of between 450°C and 550°C.

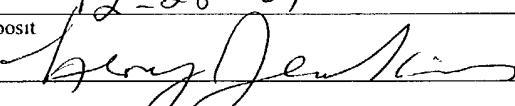
37. (New) The metathesis process as claimed in claim 35, wherein said metathesis catalyst is selected from a tungsten and molybdenum containing catalyst.

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Date of Deposit 12-28-01

Signature 

Typed or Printed Name of Person Signing Certificate Jerry Jenkins

38. (New) The metathesis process as claimed in claim 37, wherein said process is metathesis catalyst is selected from a WO_3 and a MoO_3 catalyst.

39. (New) The metathesis process as claimed in claim 35, wherein said Fischer-Tropsch olefinic feedstock in the C_5 to C_{15} range includes at least linear alpha olefins and mono-methyl branched olefins.

40. (New) The metathesis process as claimed in claim 35, wherein said Fischer-Tropsch olefinic feedstock includes one or more olefins selected from the C_5 to C_9 range.

41. (New) The metathesis process as claimed in claim 35, wherein the product of the high temperature metathesis process is used in the production of alkyl benzene, plasticizers, detergents, and/or drilling fluids, having both a linear fraction and a branched fraction.

42. (New) The metathesis process as claimed in claim 41, wherein the branched fraction is mono-methyl branched and optionally includes di-methyl, and/or ethyl branching.

43. (New) A metathesis process for the metathesis of olefins in the C_5 to C_{15} range, said metathesis process including the step of subjecting an olefinic feedstock in the C_5 to C_{15} range to a metathesis catalyst at a temperature of from 300°C to 600°C and a pressure of from 1 to 30 Bar, the process including the recycling of a part of the product of the metathesis reaction to the reaction to increase the selectivity for a desired product range.

44. (New) A metathesis process as claimed in claim 43, wherein the olefinic feedstock is a Fischer-Tropsch olefinic feedstock including mono-methyl branched olefins.

45. (New) A metathesis process as claimed in claim 44, wherein the olefinic feedstock includes one or more olefins in the C_5 to C_9 range.

46. (New) A metathesis process as claimed in claim 43, wherein the process includes a separation stage wherein a recycle fraction in the C₅ to C₈ range is separated from the product and recycled to the reaction.

47. (New) A metathesis process as claimed in claim 43, wherein the quantity of recycle in the feedstock is selected to provide a C₉ and higher selectivity of above 50%.

48. (New) A metathesis process as claimed in claim 43, wherein the recycle makes up between 20% and 80% of the reaction feedstock.

49. (New) A metathesis process as claimed in claim 48, wherein the recycle makes up between about a third and three quarters of the reaction feedstock.

50. (New) A metathesis process product composition produced by a process as claimed in claim 44, wherein the ratio of linear to branched metathesis process products is greater than 1:1.

51. (New) A metathesis process product composition as claimed in claim 50, wherein the ratio of linear to branched metathesis process products is greater than 2:1.

52. (New) A metathesis process product composition as claimed in claim 50, wherein the ratio of linear to branched metathesis process products is about 3:1.

53. (New) A metathesis process product composition as claimed in claim 50, wherein the branching of the metathesis process products is predominantly mono-methyl branching.

54. (New) A metathesis process product composition as claimed in claim 50, wherein the branching of the metathesis process products includes di-methyl and/or ethyl branching.

Applicant : Jan Mattheus Botha, et al.
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55. (New) A metathesis process product composition as claimed in claim 50, which is used in the production of alkyl benzene, plasticizers, detergents, and/or drilling fluids, having both a linear fraction and a branched fraction with the ratio of linear to branched fractions being related to the ratio of linear to branched metathesis process product composition used in their production.

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002001 / USP/F221/JL/sg

REMARKS

All amendments have been made to remove multiple dependency while conserving the claimed subject matter. No new matter has been introduced.

Attached is a marked-up version of the changes being made by the current amendment.

Claims 35-55 are now pending. Applicant submits that all of the claims are now in condition for examination, which action is requested. Please apply any charges or credits to Deposit Account No. 06-1050, referencing attorney docket no. 13777-002001.

Respectfully submitted,

Date: December 28, 2001

for 
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Reg. No. 34,053

Jeffrey D. Hsi
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Applicant : Jan Mattheus Botha, et al.
Serial No. : Unassigned
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Page : 6

Attorney's Docket No.: 13777-

002001 / USP/F221/L/kg

10/019471

531 Rec'd PCT/EP 28 DEC 2001

Version with markings to show changes made

In the claims:

Claims 1-34 have been cancelled.

HIGH TEMPERATURE METATHESIS PROCESS

531 Rec'd PCT/PT

28 DEC 2001

Field of the Invention

5 This invention relates to a high temperature metathesis process. In particular, the invention relates to the optimisation of the high temperature metathesis process to improve selectivity for a desired product range.

Background to the Invention

10

The applicant is aware that olefins in the C₉ to C₁₄ range may be used as detergent and plasticizer precursors as well as for alkylation of benzene, and that C₁₅ to C₁₈ olefin ranges may be used as drilling fluids and drilling fluid precursors, amongst other uses.

15

Conventional thinking was that linear olefins may be used to produce linear alkyl benzene and linear oxo-alcohols which could be used to produce detergents and plasticizers which were believed to be both bio-degradable and suitable for their intended purpose. Thus, previously efforts were 20 concentrated on producing linear oxo-alcohols and lineal alkyl benzene, and thus efforts were focused on linear olefins from which these could be made.

Recently, however, a new wave of thinking has lead to the belief that non-linear oxo-alcohols as well as non-linear alkyl chain alkyl benzene could 25 be used alone or together with their linear counterparts for the production of

said detergents and plasticizers. In particular short chain branched olefins are believed best suited to produce such non-linear products. Thus, recent efforts have concentrated on the delinearization of the linear olefins in order to use such olefins in the production of the non-linear products.

5

Summary of the Invention

Surprisingly, after extensive research, the applicant has found that a peculiar olefin composition in the C₉ to C₁₈ range, having both linear and non-
10 linear olefins may be made by metathesis of Fischer-Tropsch olefins in the C₅ to C₁₅ range.

Thus, according to a first aspect of the invention, there is provided a high temperature metathesis process for the metathesis of Fischer-Tropsch
15 olefins in the C₅ to C₁₅ range, said metathesis process including the step of subjecting a Fischer-Tropsch olefin feedstock in the C₅ to C₁₅ range to metathesis reaction conditions, said olefin feedstock including mono-methyl branched olefins.

20 The high temperature metathesis process may be carried out at a temperature of between 300°C to 600°C.

Typically the high temperature metathesis process is carried out at a temperature of between 450°C and 550°C.

The operating pressure of the high temperature metathesis process may be between 1 and 30 bar, or even higher.

The high temperature metathesis process may use a tungsten or 5 molybdenum based catalyst, for example, WO_3 or MoO_3 , supported or unsupported, with or without co-catalysts. The support can typically be SiO_2 , Al_2O_3 , ZrO_2 , TiO_2 , or mixtures thereof.

The high temperature metathesis process Fischer-Tropsch olefinic 10 feedstock in the C_5 to C_{15} range may include linear alpha olefins, mono-methyl branched olefins, paraffins, dienes, aromatics, and the like.

Typically, the Fischer-Tropsch olefinic feedstock includes one or more olefins selected from the C_5 to C_9 range.

15

The product of the high temperature metathesis process may include one or more mono-methyl branched olefins in the C_9 to C_{18} range.

The product of the high temperature metathesis process may include 20 one or more linear olefins in the C_9 to C_{18} range.

The product of the high temperature metathesis process may include one or more mono-methyl branched olefins and one or more linear olefins in the C_9 to C_{18} range. The olefins of the product may be internal olefins.

25

The product of the high temperature metathesis process may be used in the production of alkyl benzene, plasticizers, detergents, drilling fluids, and the like, having both a linear fraction and a branched fraction (for alkyl benzene the alkyl chain is branched or linear).

5

Typically, the branched fraction will be mono-methyl branched. However, the branching may be di-methyl and/or ethyl.

According to a second aspect of the invention, there is provided a high 10 temperature metathesis process for the metathesis of olefins in the C₅ to C₁₅ range, said metathesis process including the step of subjecting an olefinic feedstock in the C₅ to C₁₅ range to metathesis reaction conditions, the process including the recycling of a part of the product of the metathesis reaction to the reaction to increase the selectivity for a desired product range.

15

The olefinic feedstock may be a Fischer-Tropsch olefinic feedstock including mono-methyl branched olefins.

Typically, the olefinic feedstock includes one or more olefins in the C₅ 20 to C₉ range.

Where the desired product range includes olefins in the C₉ to C₁₈ range, the process includes a separation stage wherein a recycle fraction in the C₅ to C₈ range is separated from the product and recycled to the reaction.

25

The quantity of recycle in the feedstock may be selected to provide a C₉ and higher selectivity of above 50%.

Generally, the quantity of recycle in the feedstock is selected to provide
5 a C₉ and higher selectivity of above 50%.

Typically, the recycle makes up between 20% and 80% of the reaction
feedstock.

10 Usually, the recycle makes up between about a third and three quarters
of the reaction feedstock.

The total yield of high temperature metathesis process product in the
C₉ to C₁₈ range is above 40%.

15 Typically, the total yield of high temperature metathesis process
product in the C₉ to C₁₈ range is about 50%.

20 The total feedstock conversion of the high temperature metathesis
process of the invention is typically in the range of 60% to 90%, usually about
80%.

25 The ratio of linear to branched high temperature metathesis process
products is typically greater than 1:1.

Usually, the ratio of linear to branched high temperature metathesis process products is greater than 2:1.

Generally, the ratio of linear to branched high temperature metathesis process products is about 3:1.

The branching of the high temperature metathesis process products is predominantly mono-methyl branching, although some di-methyl, and/or ethyl branching may also be present.

10

The product of the high temperature metathesis process may be used in the production of alkyl benzene, plasticizers, detergents, drilling fluids, and the like, having both a linear fraction and a branched fraction (for alkyl benzene the alkyl chain is branched or linear), the ratio of linear to branched fractions being related to the ratio of linear to branched high temperature metathesis process products used in their production.

Description of the Drawing and Examples

20 The invention will now be described, by way of non-limiting illustration only, with reference to the accompanying line diagram.

In the diagram, reference numeral 10 generally indicates a high temperature metathesis process broadly in accordance with the invention.

25

The process 10 includes a reactor 12 operated at between 450°C and 550°C and at an operating pressure of between 1 and 30 bar. A Fischer-Tropsch olefinic feedstock 14 including mono-methyl branched olefins, is fed into the reactor 12. The feedstock 14 includes olefins in the C₅ to C₉ range.

5

Usually the feedstock 14 will be purified of oxygenates which may poison the catalyst by extractive distillation (not shown), prior to being fed to the reactor 12.

10

The reaction product 16 includes both linear and branched internal olefins in the C₂ to C₁₈ range.

The reaction product 16 is fed to a separator 18 where it is cut into a light product stream 20 including C₂ to C₄, a recycle stream 22 including C₅ to 15 C₈, and a heavy product 24 including product in the desired C₉ to C₁₈ range.

The recycle stream 22 is combined with the feedstock 14 to form the total feedstock of the reactor 12.

20

The recycle stream 22 is between a third and three quarters of the feedstock 14.

25

The total yield of heavy product stream 24 is about 50%, while the feedstream 14 conversion is about 80%, with a selectivity for C₉ to C₁₈ of about 60%.

The ratio of linear to branched product in heavy product stream 24 is about 3:1

5

Examples

Several runs were made by passing olefin containing feed downwards through a vertical pipe reactor, unless otherwise stated. This reactor (25.4 mm in diameter and 400 mm in length) was positioned in a temperature-controlled electric furnace with a thermocouple positioned in the catalyst bed to monitor reaction temperatures.

About 100 mm depth of glass beads (2 mm diameter) were placed at the bottom of the pipe reactor supported by a layer of quartz wool. Another layer of quartz wool was placed on top of the glass beads as support for the catalyst bed comprising of about 12 g of catalyst. This was topped with another layer of quartz wool and the remainder of the reactor filled with glass beads. The catalyst was activated by heating at 550°C in flowing air for 12 hours, followed by heating at 600°C for 2 hours under a flow of nitrogen and finally the catalyst was cooled under a flow of nitrogen to reaction temperature (typically 500°C).

Example 1

In this Example a catalyst in the form of a WO₃ supported on SiO₂ was used, in which the WO₃ and SiO₂ were in a mass ratio of 8:92. The process

was operated in the temperature range of 400 to 550°C and at a LHSV of 1 h⁻¹. As a feed was used a C₇ SLO narrow cut after NMP extraction, containing 3-methyl-1-hexene (0.7870%), 5-methyl-1-hexene (1.9068%), 4-methyl-1-hexene (3.1737%), 2-methyl-1-hexene (4.1847%), 2-methylhexane (1.6501%), 3-methylhexane (2.8000%), 1-heptene (74.5710%), n-heptane (6.3012%), 2-methyl-2-hexene (0.6832%), 3-heptene (0.3163%), 2-heptene (0.7038%) and dienes, cyclic olefins and aromatics (2.4386%) amongst others, based on mass% calculations. Results are set forth in the following table, Table 1:

10

Table 1

Temp °C	400	450	475	500	525	550
C ₇ Conversion (%)	4.4	20.4	50.0	65.9	71.9	78.4
Yield C ₉ – C ₁₄ (%)	2.4	8.9	20.3	23.9	20.2	13.9
Selectivity C ₉ – C ₁₄ (%)	55.6	43.9	40.6	36.3	28.1	17.7
Selectivity C ₂	0.4	0.3	0.3	0.7	1.2	2.7
Selectivity C ₃	5.0	2.6	2.7	4.6	7.7	14.0

15

Example 2

In this Example a catalyst in form of a WO₃ supported on SiO₂ was used, in which the WO₃ and SiO₂ were in a mass ratio of 8:92. The process was operated at 500°C and by recycling some of the olefins formed back to the reactor. As a feed was used a C₇ SLO narrow cut after NMP extraction, containing 3-methyl-1-hexene (0.7870%), 5-methyl-1-hexene (1.9068%), 4-methyl-1-hexene (3.1737%), 2-methyl-1-hexene (4.1847%), 2-methylhexane

(1.6501%), 3-methylhexane (2.8000%), 1-heptene (74.5710%), n-heptane (6.3012%), 2-methyl-2-hexene (0.3163%), 2-heptene (0.7038%) and dienes, cyclic olefins and aromatics (2.4386%) amongst others, based on mass% calculations. Results are set forth in the following table, Table 2:

5

Table 2

Run	Feed Conversion (%)	C ₈ Yield (%)	C ₉ – C ₁₀ Yield (%)	C ₁₁ – C ₁₄ Yield (%)	C ₁₅ – C ₁₈ Yield (%)	C ₈ – C ₁₄ Yield (%)
1 ^a	89.7	4.8	7.0	36.5	4.0	48.3
2 ^b	96.4	2.1	22.1	33.5	5.5	57.7
3 ^c	90.6	4.6	33.0	27.1	0.5	64.7
4 ^d	90.1	11.8	31.3	22.8	0.2	65.9

(a) 1.0 LHSV based on fresh feed; 6.0 LHSV with recycle (1:5 recycle ratio);
10 (Recycle C₅ – C₁₀)

(b) 1.4 LHSV based on fresh feed; 5.6 LHSV with recycle (1:3 recycle ratio);
15 (Recycle C₅ – C₉)

(c) 1.4 LHSV based on fresh feed; 5.6 LHSV with recycle 1:3 recycle ratio;
15 (Recycle C_{5/6} – C₈)

(d) 2.0 LHSV based on fresh feed; 5.0 LHSV with recycle 1:1.5 recycle ratio;
15 (Recycle C_{4/5} – C₇)

Example 3

20 In this Example a catalyst in the form of a WO₃ supported on SiO₂ were in a mass ratio of 8:92. The process was operated at 500°C and at a LHSV of 3 h⁻¹. As a feed was used a C₅ SLO co-monomer grade cut containing 99% 1-pentene. The C₅ – C₇ fraction was recycled (1:1 recycle ratio) back to the

reactor in order to increase the yield towards the C₈ – C₁₄ fraction. Results are set forth in the following table, Table 3:

Table 3

5

Temp °C	500
C₅ Conversion (%)	88.2
Yield C₉ – C₁₄ (%)	19.9
Selectivity C₉ – C₁₄ (%)	22.6
Selectivity C₂	5.2
Selectivity C₃	19.4

The applicant believes that it is an advantage of the invention as illustrated, that the high operating temperatures result in a high degree of 10 resistance to poisoning of the metathesis catalyst by feedstock components, such as branched olefins, dienes, aromatics, and the like.

The applicant believes that it is a further advantage of the invention as illustrated that by recycling a cut of the product which is below the desirable 15 carbon length range, high selectivity to desired products is achieved..

Claims:

1. A high temperature metathesis process for the metathesis of Fischer-Tropsch olefins in the C₅ to C₁₅ range, said metathesis process including the step of subjecting a Fischer-Tropsch olefin feedstock in the C₅ to C₁₅ range to metathesis reaction conditions, said olefin feedstock including mono-methyl branched olefins.
2. The high temperature metathesis process as claimed in claim 1, wherein said process is carried out at a temperature of between 300°C to 600°C.
3. The high temperature metathesis process as claimed in claim 1, wherein said process is carried out at a temperature of between 450°C and 550°C.
4. The high temperature metathesis process as claimed in any one of claims 1 to 3, wherein said process is carried out at a pressure of between 1 and 30 bar.
5. The high temperature metathesis process as claimed in any one of claims 1 to 4, wherein said process is carried out in the presence of a tungsten or molybdenum based catalyst.

6. The high temperature metathesis process as claimed in any one of claims 1 to 4, wherein said process is carried out in the presence of a WO_3 or MoO_3 catalyst.

5 7. The high temperature metathesis process as claimed in any one of the preceding claims, wherein said Fischer-Tropsch olefinic feedstock in the C_5 to C_{15} range includes at least linear alpha olefins and mono-methyl branched olefins.

10 8. The high temperature metathesis process as claimed in any one of the preceding claims, wherein said Fischer-Tropsch olefinic feedstock includes one or more olefins selected from the C_5 to C_9 range.

9. The high temperature metathesis process as claimed in any one 15 of the preceding claims, wherein the product of the high temperature metathesis process includes one or more mono-methyl branched olefins in the C_9 to C_{18} range.

10. The high temperature metathesis process as claimed in any one 20 of the preceding claims, wherein the product of the high temperature metathesis process includes one or more linear olefins in the C_9 to C_{18} range.

11. The high temperature metathesis process as claimed in any one of the preceding claims, wherein the olefins of the product are internal olefins.

12. The high temperature metathesis process as claimed in any one of the preceding claims, wherein the product of the high temperature metathesis process is used in the production of alkyl benzene, plasticizers, detergents, and/or drilling fluids, having both a linear fraction and a branched fraction.

13. The high temperature metathesis process as claimed in claim 12, wherein the branched fraction is mono-methyl branched.

10 14. The high temperature metathesis process as claimed in claim 13, wherein the branched fraction includes di-methyl, and/or ethyl branching.

15. A high temperature metathesis process for the metathesis of olefins in the C₅ to C₁₅ range, said metathesis process including the step of 15 subjecting an olefinic feedstock in the C₅ to C₁₅ range to metathesis reaction conditions, the process including the recycling of a part of the product of the metathesis reaction to the reaction to increase the selectivity for a desired product range.

20 16. A high temperature metathesis process as claimed in claim 15, wherein the olefinic feedstock is a Fischer-Tropsch olefinic feedstock including mono-methyl branched olefins.

17. A high temperature metathesis process as claimed in claim 15 or claim 16, wherein the olefinic feedstock includes one or more olefins in the C₅ to C₉ range.

5 18. A high temperature metathesis process as claimed in claim 15, wherein the desired product range includes olefins in the C₉ to C₁₈ range.

10 19. A high temperature metathesis process as claimed in claim 18 wherein the process includes a separation stage wherein a recycle fraction in the C₅ to C₈ range is separated from the product and recycled to the reaction.

15 20. A high temperature metathesis process as claimed in claim 19, wherein the quantity of recycle in the feedstock is selected to provide a C₉ and higher selectivity of above 50%.

21. A high temperature metathesis process as claimed in claim 19 or claim 20, wherein the recycle makes up between 20% and 80% of the reaction feedstock.

20 22. A high temperature metathesis process as claimed in claim 21, wherein the recycle makes up between about a third and three quarters of the reaction feedstock.

23. A high temperature metathesis process as claimed in any one of claim 18 to 22, wherein the total yield of high temperature metathesis process product in the C₉ to C₁₈ range is above 40%.

5 24. A high temperature metathesis process as claimed in any one of claim 18 to 22, wherein the total yield of high temperature metathesis process product in the C₉ to C₁₈ range is above 50%.

10 25. A high temperature metathesis process as claimed in any one of claims 15 to 24, wherein the total feedstock conversion is in the range of 60% to 90%.

15 26. A high temperature metathesis process as claimed in claim 25, wherein the total feedstock conversion is about 80%.

27. A high temperature metathesis process as claimed in any one of claims 15 to 26, wherein the ratio of linear to branched high temperature metathesis process products is greater than 1:1.

20 28. A high temperature metathesis process as claimed in any one of claims 15 to 27, wherein the ratio of linear to branched high temperature metathesis process products is greater than 2:1.

29. A high temperature metathesis process as claimed in any one of claims 15 to 28, wherein the ratio of linear to branched high temperature metathesis process products is about 3:1.

5 30. A high temperature metathesis process as claimed in any one of claims 15 to 29, wherein the branching of the high temperature metathesis process products is predominantly mono-methyl branching.

10 31. A high temperature metathesis process as claimed in any one of claims 15 to 30, wherein the branching of the high temperature metathesis process products includes some di-methyl and/or ethyl branching.

15 32. A high temperature metathesis process as claimed in any one of claims 15 to 31, wherein the products of the high temperature metathesis process are used in the production of alkyl benzene, plasticizers, detergents, and/or drilling fluids, having both a linear fraction and a branched fraction with the ratio of linear to branched fractions being related to the ratio of linear to branched high temperature metathesis process products used in their production.

20

33. A high temperature metathesis process substantially as herein described and illustrated.

25 34. A new high temperature metathesis process substantially as herein described.

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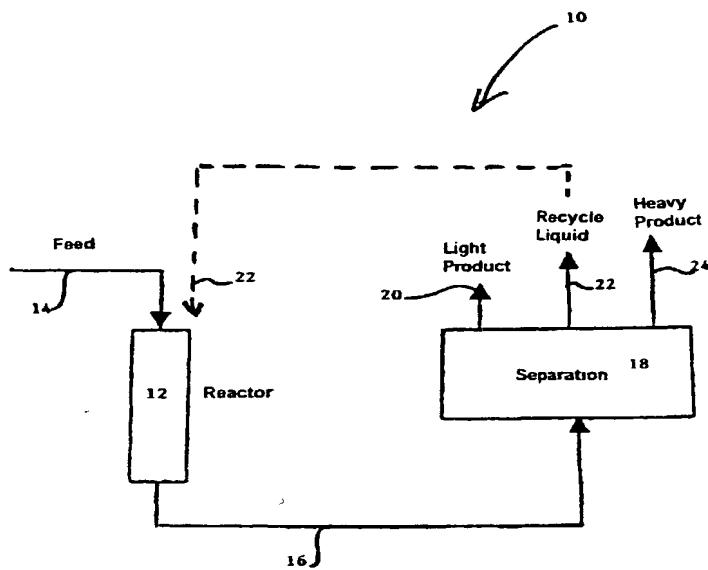
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(54) Title: HIGH TEMPERATURE METATHESIS PROCESS



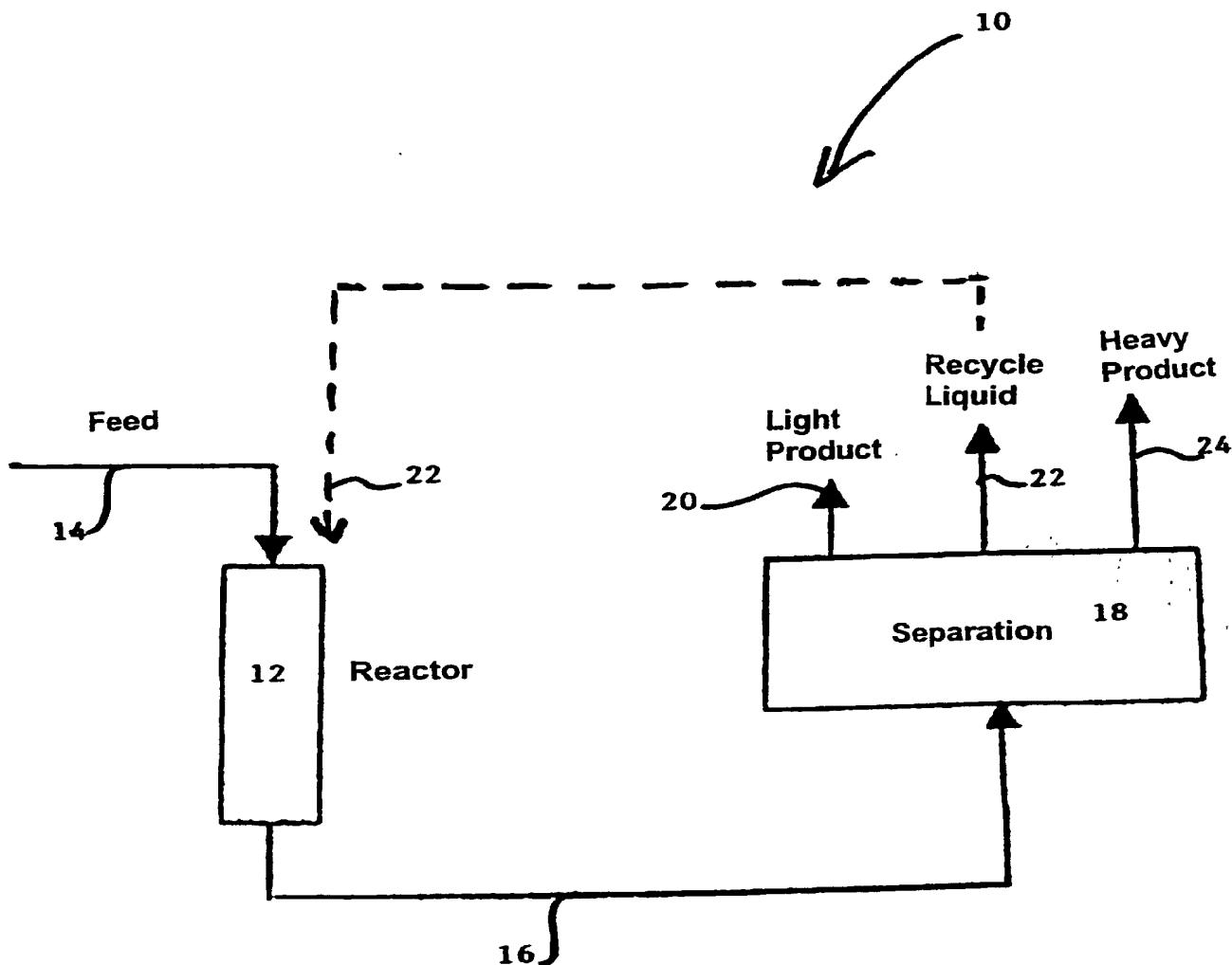
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(57) Abstract: The invention provides a high temperature metathesis process for the metathesis of Fischer-Tropsch olefins in the C₅ to C₁₅ range, said metathesis process including the step of subjecting a Fischer-Tropsch olefin feedstock in the C₅ to C₁₅ range to metathesis reaction conditions, said olefin feedstock including mono-methyl branched olefins. The invention also provides alkyl benzenes (AB's), drilling fluids and oxo-alcohols produced from the products of the metathesis process.

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COMBINED DECLARATION AND POWER OF ATTORNEY

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled **HIGH TEMPERATURE METATHESIS PROCESS**, the specification of which:

is attached hereto.
 was filed on December 28, 2001 as Application Serial No. 10/019,471 and was amended on December 28, 2001.
 was described and claimed in PCT International Application No. _____ filed on _____ and as amended under PCT Article 19 on _____.

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose all information I know to be material to patentability in accordance with Title 37, Code of Federal Regulations, §1.56.

I hereby claim the benefit under Title 35, United States Code, §119(e)(1) of any United States provisional application(s) listed below:

U.S. Serial No.	Filing Date	Status
60/142,382	July 6, 1999	Abandoned

I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose all information I know to be material to patentability as defined in Title 37, Code of Federal Regulations, §1.56(a) which became available between the filing date of the prior application and the national or PCT international filing date of this application:

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Country	Application No.	Filing Date	Priority Claimed
South Africa	ZA 99/4380	July 6, 1999	<input checked="" type="checkbox"/> Yes <input type="checkbox"/> No
WIPO	PCT/ZA00/00120	July 6, 2000	<input checked="" type="checkbox"/> Yes <input type="checkbox"/> No

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Combined Declaration and Power of Attorney

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